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" A method for effecting recombination of gases evolved during the over charging processes back into water in sealed lead acid storage cells and batteries ".

COUNCIL OF SCIENTIFIC & INDUSTRIAL RESEARCH, Rafi Marg, New Delhi-1, India, an Indian registered body incorporated under the registration of Societies Act (Act XXI of 1860).

The following specification describes the nature of this invention :-

This is an invention by Subbish Palanichamy, Scientist; Pennagaram Vyasarao Vasudeva Rao, Scientist and Handady Venkatakrishna Udupa, Director, all of Central Electrochemical Research Institute, KaraiKudi, Tamil Nadu, India, all Indian citizens.

PRICE : TWO RUPEES

This invention relates to improvements in or relating to catalysts for gas recombination in sealed storage cells and batteries.

Hitherto it has been proposed to use the storage cells and batteries with open type vent plugs or pressure vents for the escape of gases formed during charging process without recourse to a suitable process for recombining these gases again to water.

This is open to the objection that the level of the electrolyte has to be maintained to the proper mark by adding distilled water periodically and further the top of the cells and batteries have to be cleaned to minimise the corrosive action due to leakage of the electrolyte on the terminals. The cells or batteries could not be used in any position.

The object of the invention is to obviate these disadvantages by the use of a suitable catalyst capable of

recombining the gases formed during overcharging process back into water so that the need for topping up with distilled water and cleaning of the terminals are eliminated and the cells are capable of hermetic sealing making them absolutely maintenance free.

To these ends, the invention broadly consists in the use of a suitable catalyst capable of recombing hydrogen and oxygen formed during charging process to form water and the method of making such a catalyst composition for the said purpose.

The catalyst is prepared by taking suitable amount of noble metal salts such as palladium chloride, palladium nitrate or chloro platinic acid or its salts, or iridium chloride and mixing the salts either singly or in combination with catalytic carrier like silica, alumina or titania taken in suitable proportion to get a large active surface for the gas recombination reaction. The carrier employed is in the form of fine powder with a specific surface area not less than 100 m^2/gm. The carrier containing the noble metal salts is then treated in a reducing atmosphere at a temperature 150°C to 450°C to convert the noble metal salts into an active form of finely dispersed metal. The catalyst is further stabilised in its active form by using suitable stabilising agents.

The following typical examples are given to illustrate the invention:

EXAMPLE - I

Weight of the carrier	:	3 gm
Ratio of the catalyst (platinum) to carrier	:	1:333
Surface area of the carrier	:	102 m^2/g
Capacity of the cell incorporated with the catalyst	:	700 mAH
Overcharge current	:	2 mA/cm^2

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EXAMPLE - II

Weight of the carrier	: 2 gm
ratio of the catalyst (Palladium) to the carrier	: 1:500
Surface area of the carrier	: 102 m^2/g
Capacity of the cell incorporated with the catalyst	: 700 mAH
Overcharge current	: 2 mA/cm^2
Life of catalyst for 100% activity	: 200 hours

The following are among the main advantages of the invention:

1) There is no evolution of corrosive gases during charging of cells and batteries incorporated with the catalyst.

2) The cells incorporating the catalyst can be sealed completely so that they can be used in any position.

3) Maintenance procedures such as addition of distilled water, cleaning of terminas are not required.

4) The cells can be incorporated directly into the portable electrical and electronic instruments for providing electric power.

Dated This 25th day of March, 1975

Asstt. Technical Officer,
Council of Scientific & Industrial Research

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THE PATENTS ACT. 1970

COMPLETE SPECIFICATION

(Section—10)

" A method for effecting recombination of gases evolved during the over charging processes back into water in sealed lead acid storage cells and batteries."

COUNCIL OF SCIENTIFIC AND INDUSTRIAL RESEARCH, Rafi Marg, New Delhi-1, India, an Indian registered body incorporated under the Registration of Societies Act (Act XXI of 1860).

The following specification particularly describes and ascertains the nature of this invention and the manner in which it is to be performed :-

This is an invention by Subbiah Palanichamy, Scientist; Pennagaram Vyasarao Vasudeva Rao, Scientist and Handady Venkatakrishna Udupa, Director, all of Central Electrochemical Research Institute, Karaikudi-623006, Tamil Nadu, India; all Indian citizens.

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/a method for effecting recombination of gases evolved during the over charging processes back into water in sealed lead acid storage cells and batteries and has particular reference to

This invention relates to/improvements in or relating to the method of effecting gas recombination of hydrogen and oxygen gases formed during charging in sealed lead acid storage cells and batteries.

Hitherto it has been proposed to use different methods for avoiding the increase in pressure during charging in lead acid cells and batteries for achieving sealed type construction. One of the methods consists in providing a pressure sensitive valve which will allow the gases to pass to the atmosphere when the pressure of the accumulated gas exceeds a predetermined level. Another method consists in using catalytic electrodes capable of recombining the gases electrochemically into water. A third method suggested was to use catalysts based on noble metals.

The draw back connected with the above methods are either that the cells cannot be sealed completely or the life of the cells and storage batteries incorporating the above methods is limited.

The object of the invention is to describe a method of effecting recombination of gases formed during over charging process back into water and a process therefor so that the need for topping up with distilled water and cleaning of the terminals are eliminated and the cells are capable of hermetic sealing, thus making the cells absolutely maintenance free.

Among the methods suggested, the method based on the use of catalyst appears to be simple for the above application. The efficiency and life of the catalyst depend upon the purity of raw materials, particle size of the carrier and the amount of the noble metal present. For example, in one of the patents, it is disclosed that use of 0.01% Palladium dispersed in 0.25 inch dia and 0.375 inch length with a specific surface area $<1\ m^2/g$ produced a temperature of 260°C at a gassing produced from 4 Amps.

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The object of this invention is to describe a method for effecting gas recombination the gases formed during overcharging process back into water using catalyst material and a process therefor so that the need for topping up with distilled water and cleaning of the terminals are eliminated and the cells are capable of hermetic sealing, thus making the cells and batteries maintenance free.

The invention broadly consists of noble metals suitably dispersed over inert catalyst carrier. The catalyst carrier must be free from any impurities like manganese arsenic or sulphur which are harmful. It was observed that the performance of the catalyst was improved when the surface area of the catalyst used was above 100 m^2/g preferably in the range of 102 m^2/g and 130 m^2/g.

In the conventional type lead acid batteries, the gases formed during the over charge is allowed to escape to the atmosphere. This is objectionable if such batteries are used for supplying electrical energy to portable electrical and electronic equipments.

The said catalyst is prepared by taking suitable amount of noble metal salts such as palladium chloride, palladium nitrate, chloroplatinic acid and mixing with the catalyst carrier. The mass mixed thoroughly to get a uniform composition and metal salt is converted to an active form by heating in a reducing atmosphere containing hydrogen at a temperature of 150-450°C. After the reduction the mass is cooled to room temperature. The mass is ground to get a fine powder. The catalyst is then given a stabilising treatment by passing purified oxygen at a temperature of 35 - 90°C for a period ranging between 30 minutes to 2 hours. Three grams of the resultant catalyst was found to be active for a period

of 120 hours when a lead acid cell of 700 mAH capacity was over charged at a current density of 2 mA/cm^2.

The present invention consists of a method for effecting gas recombination in sealed lead acid storage cells and batteries and a process therefor which comprises a noble metal suitably dispersed over inert catalyst carrier like alumina, silica, or titania in suitable proportions and stabilising treatment to keep the noble metal in its active form.

The following are the typical examples illustrating the invention.

Example 1

3 gm of alumina powder having a specific surface of 102 m^2/g is treated with enough metal salt solution (e.g) chloroplatinic acid to give noble metal and catalyst carrier ratio of 1:333 on weight basis. The mass is stirred well to get a uniform composition. Then it is treated with Hydrogen gas at a temperature of 150 - 250°C, preferably at 175°C for a period of 4 hours. The mass is cooled and ground into powder.

The above powder when tested in a 2 volt 700 mA hr lead acid cell was found to be active for only 45 hours for the recombination reaction at overcharge current density of 2 mA/cm^2.

Example 2

Same as in example 1 but the catalyst material was given an activation/stabilising treatment by passing purified oxygen at a temperature of 35-90°C preferably at 40°C for a period of 1 hour. The activity of the catalyst was found to be increased to 85 hours.

Example 3

2 gm of alumina powder having a specific surface of 102 m^2/g is treated with noble metal salt such as palladium chloride containing enough noble metal to give a ratio of 1:500 of the weight of the carrier. The mass is stirred well to get a uniform composition. The mass is treated with purified hydrogen gas at a temperature of 175°C for a period of 4 hours. The mass is cooled to room temperature and ground to a powder form. The catalyst material was given a activation/stabilising treatment in a stream of oxygen at temperature of 40°C for a period of 1 hour. The activity of the above catalyst was found to be 200 hours when tested in a 2 m volt lead acid cell having a nominal capacity of 700 mAH, when the cell was over charged at current density of 2 mA/cm^2.

Example 4

Same as in example 3 except that the specific surface of the alumina powder was 85 m^2/g. The activity of the above catalyst was found to be 98 hours only.

The advantage of this method is that there is no evolution of gases during charging of cells and batteries incorporated with the catalyst.

The cells incorporating the catalyst can be sealed completely so that they can be used in ar. position.

Maintenance procedure such as addition of distilled water, cleaning of terminals are not required.

The cells can be incorporated directly into the portable electrical and electronic instruments for providing electric

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Summary

An invention is described for a method for recombining oxygen and hydrogen gases formed during over charge of lead acid storage cells and batteries back into water and a process therefor while the use of lead acid battery system for automobiles, standby power and traction is well established, this system has not entered into the field of sealed type batteries. This is due to the fact that during normal charging and over charging, hydrogen and oxygen gases are evolved, the rate of gas evolution being proportional to the charging current. In the absence of any gas recombination device, the pressure inside the cell box increases with time considerably thus making any attempt to seal the cell very difficult.

The invention consists in the preparation of catalyst composition based on noble metals such as platinum and/or palladium suitably dispersed over inert carrier like alumina. The catalyst is further given a stabilising treatment to keep the noble metal in its active form.

We **Claim**:

1) A method for effecting recombination of gases evolved during the over charging processes back into water in sealed lead acid storage cells and batteries by the use of a catalyst capable of recombining the gases characterised in that a noble metal such as platinum or palladium dispersed over inert carriers' such as alumina, silica and titania activated by treatment with purified hydrogen gas and further stabilised by treatment wi' stream of oxygen, is used as catalyst.

2) A process as claimed in claim (1) wherein the activating treatment is given by passing purified hydrogen gas at a temperature of 150°C to 450°C preferably 175°C for a period ranging from 1 hour to 4 hours.

3) A process as claimed in claims 1 and 2 wherein the stabilising treatment is given by passing a stream of oxygen at a temperature of 35-90°C preferably at 40°C for a period of 30 minutes to 1 hour.

Dated this 1st day of July, 1976.

R Bhaskar Rai

PATENT OFFICER,
Council of Scientific & Industrial Research.